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STONEMAN II TEST OF RECLAMATION PERFORMANCE

VOLUME I

THE PRODUCTION, DISPERSAL AND
MEASUREMENT OF SYNTHETIC FALLOUT MATERIAL

Research and Development Technical Report USNRDL-TR-334

6 June 1960

by

W. B. Lane
J. D. Sartor



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U.S. NAVAL RADIOLOGICAL DEFENSE LABORATORY

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ABSTRACT

A field test was conducted in the Fall of 1958 wherein multicurie amounts of radioisotopes were used to prepare ton quantities of synthetic fallout for the evaluation of land reclamation procedures. The processing of the radioisotope La^{140} , the procedures for radio-tagging clay soil, and the measuring of gamma radiation from large contaminated areas are described. The factor for converting gamma radiation measurements to equivalent weights of soil was determined so that decontamination effectiveness could be expressed in mass units.

SUMMARY

The Problem

Ton quantities of synthetic fallout material are required for the evaluation of the cost and performance of land target recovery procedures. Methods of producing and dispersing the synthetic fallout material had to be developed and also a factor for converting gamma radiation measurements to equivalent weights of soil had to be determined.

Findings

Operation Stoneman II was conducted during August and September 1958. This report contains a description of the methods used in preparing ton quantities of synthetic fallout material used in the Operation. Production facilities which included a hot cell, isotope preparation apparatus and radio-tagging equipment were designed and fabricated for the manufacture of ton quantities of synthetic fallout material. Analytical techniques for the determination of both mass distribution and radioactivity distribution as a function of particle size were developed. A factor for the conversion of gamma radiation measurements to equivalent weights of soil was determined.

ADMINISTRATIVE INFORMATION

This investigation, sponsored by the Department of the Army, was done as part of Program B-3, Problem 3, described in this Laboratory's USNRDL Technical Program for Fiscal Year 1959, revised 1 January 1959. Specifically, the work was based on objective (1): "The production and evaluation of a simulant of fallout from nuclear detonations on land, and the determination of contamination-decontamination between simulated fallout and land target surfaces."

The other volumes in this series of reports are:

- Vol. II Performance Characteristics of Wet
 Decontamination Procedures
- Vol. III Performance Characteristics of Dry
 Decontamination Procedures
- Vol. IV Performance Characteristics of Land
 Reclamation Procedures
- Vol. V Some Contaminability Characteristics
 of Personnel Exposed to Contact Beta
 Radiation

ACKNOWLEDGEMENTS

The objectives of this test could not have been fulfilled without the whole-hearted assistance and cooperation of many organizations and personnel therefrom. The performance of the 50th Chemical Service Platoon, U.S. Army, assigned to support the test was outstanding in every respect. The services of the personnel from the Mobile Construction Battalion Five, Port Hueneme, California, proved invaluable in the operation of heavy equipment assigned to the project. In addition the authors wish to acknowledge the invaluable aid from the following organizations: Headquarters, Sixth U.S. Army, Presidio of San Francisco, California; Post Engineer, Camp Stoneman, California; Research Directorate, Air Force Special Weapons Center, Kirtland Air Force Base, New Mexico; U.S. Naval Civil Engineering Laboratory, Port Hueneme, California.

The individual contributions to the synthetic fallout design and preparation phases by L.W. Weisbecker, presently of Aerojet Nucleonics, San Ramon, California, and of J.W. Schulte of Los Alamos Scientific Laboratory are acknowledged.

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CHAPTER 1

INTRODUCTION

1.1 OBJECTIVE

The Stoneman II Tests of Reclamation Performance investigated a variety of proposed decontamination procedures. This report describes the experimental and production phases of the manufacture and dispersal of ton quantities of synthetic radioactive fallout which were produced for the contamination-decontamination studies. Some fundamental concepts and theory are presented, but the practical operational aspects involved in the field test are emphasized.

1.2 BACKGROUND

Passive defense against nuclear weapons includes the removal of radioactive fallout which originates from nuclear detonations, and the need for effective countermeasures for the protection and radiological recovery of land targets has long been apparent. The countermeasures described in NavDocks TP-PL-13, "Radiological Recovery of Fixed Military Installations", are based on limited experimental data, and additional input information is required to assure comprehensive, sound operational planning guides to enlarge the scope and increase the usefulness of that document.

The composition and properties of the radioactive fallout depends upon the type of weapon, and the nature of the surface over which or under which the detonation takes place. These debris components can be grouped as fission products, bomb products, and environmental products, and the latter two can be represented by induced radionuclides, as well as nonactive carrier materials. If the nuclear detonation occurs in sea water, the contaminant may arrive as liquid droplets in which many of the radioactive fission products are in the ionic form and therefore free to react directly with the surface material. If the detonation takes place on or under a land surface, then the properties

of the fallout will be largely controlled by the physical and chemical composition of the environmental carrier materials, since the radionuclides will be condensed on or in heated soil particles. Mixing is most intimate in fused particles from a land surface explosion and a lesser amount are adsorbed to the surface of particles which were not subjected to melting temperatures.

Decontamination involves removing the radioactive particles from the contaminated surface and transporting them to a disposal area, so that their radiation will not contribute to the dose rate within the cleaned area. The obvious reason for decontamination is to reduce the biological hazard, which is in turn related directly to reducing the radiation dose. Therefore, to be most meaningful, the decontamination effectiveness of a countermeasure must be evaluated as a ratio of the dose from the decontaminated surface to the dose which would have occurred had the surface not been decontaminated. Dose rate measurements (counts/min, mr/hr, etc.) are widely used to obtain decontamination data; however, a simple ratio of dose rates, before and after decontamination, does not always meet the requirements for a measure of decontamination effectiveness as noted above.

If the decontamination method selectively removes certain of the radioactive elements and leaves others, the decay rate of the remaining contaminant will not be the same as the decay rate of the original contaminant; therefore the radiation dose at any given time will reflect this difference. This selective removal or fractionation was demonstrated for ionic fallout from a seawater detonation.¹ However, in dry fallout, the radioactive atoms are either fused in, or strongly adsorbed to, soil particles. No data is available which shows that these adsorbed atoms are desorbed or removed from the soil particles by liquid decontamination methods. As it is unlikely that the radionuclides would enter into separate interactions with the surface, decontamination involves physically removing the fallout and transporting it away. The ratio of the mass of material remaining after decontamination to the mass of material initially deposited is equal to the ratio of the dose rate remaining to the dose rate initially present. Also since the decay of the decontaminated surface is the same as the decay of the nondecontaminated surface, the ratio of the dose rates is the same as the ratio of the doses at any time after detonation.

The material covered in this report deals exclusively with dry synthetic fallout. It is assumed that the radionuclides are fixed to the soil particles and are not desorbed by the decontamination procedures, and that radioactivity is removed to the extent the mass of soil is removed. A comprehensive presentation of the theory of radiological decontamination can be found in Miller's "Theory of

Decontamination - Part 1",² where in addition to a mathematical treatment of contamination and decontamination behavior, a logical system of terminology and notation is proposed, and a consistent technical basis is presented for interpreting data from decontamination experiments.

1.3 HISTORY

Decontamination tests were conducted at the Nevada test site during the "U" shot of Operation JANGLE, but the measurements were limited to structures and surfaces at only one location in the fallout pattern. The tests were hampered by unsuitable weather and other unpredictable conditions which so often attend the field testing of nuclear weapons, and the results were useful primarily as guide lines to describe the problem. Obvious advantages are inherent in the use of a fallout simulant in controlled experiments, both in the field and laboratory, to study decontamination reactions and develop countermeasures. This approach was initiated in a field test conducted at the U.S. Naval Advance Base Personnel Depot, San Bruno, Calif., utilizing both liquid and slurry contaminants traced with the beta-emitting radioisotope Y^{90} . Again much was learned about the experimental design for decontamination studies, but it was soon evident that data based on beta measurements could not be corrected for self-absorption and scattering.

The present theory of contamination and decontamination was developed on somewhat the same time scale but rather independently from the experimental program. Miller's fundamental concepts of decontamination mechanisms made possible the quantitative expression of important and needed experimental parameters. Thus the mass-radiation ratio was defined as the mass of material falling on a unit area to give a unit dose rate at one hour after detonation, with the dimensions of mg/sq ft/r/hr at 1 hr. Evaluation of this ratio makes possible the study of specified contaminating events by using fallout mass levels that can be simulated through the required amount of inert carrier material being deposited on a surface. Decontamination procedures can then be applied to that surface, and the amount of mass removed becomes a measure of the decontamination effectiveness. In field tests these initial and residual mass measurements would be quite impossible to make by conventional weighing methods; however, the measurements can be conveniently and accurately made by gamma-emitting radioisotope tracer techniques. The amount or level of radioisotopes is determined by the gamma radiation necessary for instrument response and has nothing whatsoever to do with the radiation levels described in the contaminating event. This and other concepts concerning

the physical and chemical nature of the fallout from high and low yield weapons, and specific designations of contaminating events which require countermeasure application, were incorporated into the design of another field test, designated as Stoneman I.^{3,4}

1.4 SUMMARY OF SYNTHETIC FALLOUT PREPARATION FOR STONEMAN I

The experiment* was conducted on the site of the inactivated Army Base, Camp Stoneman, near Pittsburgh, California, in the fall of 1956, as a joint effort of the Army Chemical Corps, Bureau of Yards and Docks, and USNRDL. The tests were designed to study the following conditions; contaminated areas of 1000 and 10,000 r/hr at 1 hr, a dry fallout from a low yield (KT) burst or a high yield (MT) land or shallow water burst, and a wet slurry fallout from a low yield (KT) shallow water burst. The mass levels used were based on a mass-radiation ratio of 25 mg/sq ft/r/hr at 1 hr. Depending on the areas to be contaminated, 1 to 2 tons of synthetic fallout were produced per day. The environmental material selected to simulate the carrier component of harbor fallout was mud dredged from San Francisco Bay, and that for the dry fallout was soil from Camp Stoneman area, which was characterized by USDA as Ambrose clay loam. Fifty tons of each of these materials were taken to the South Pacific Division Laboratory, Corps of Engineers, Sausalito, California, for shredding, drying, crushing, and screening. Material retained on a 30 mesh sieve was rejected and all the rest was used in the simulant batching without further separation. A motor pool building at Camp Stoneman was converted to an isotope processing facility. Concrete blocks were placed to form a 3 ft thick shielding wall, closed on three sides with one end open. A 2 by 2 by 2 ft zinc bromide viewing window was set in the front face of the wall, and a pair of Argonne Model 8 master-slave manipulators was provided for remote handling.

The radioisotope La^{140} was chosen as the tracer for several reasons: its gamma radiation was adequate for good instrumentation;** its 40-hr half-life was short enough to die out in a reasonable time and yet long enough to permit time tolerances in decontamination operations; it was readily produced as a very pure radiochemical salt by neutron irradiation of La^{139}Cl ; and its trivalent ion was strongly adsorbed on Ambrose clay loam.⁴

The tracer-tagged simulant was prepared as follows. Quartz capsules, each containing 2 g of lanthanum chloride, were irradiated

* An account of the test procedures and decontamination results are reported in Reference 3.

** Approximately 0.25 mc of La^{140} were required per sq ft of area to give survey instruments (T1B) readings of 15 to 30 mr/hr at 3 ft.

in the Material Testing Reactor at Arco, Idaho and placed in lead shipping containers. Containers were flown from Idaho Falls to Travis Air Force Base, Calif. in a USAF C-47 aircraft. Three flights were made per week carrying two pigs per flight. The isotopes were transported to Camp Stoneman by Army truck and unloaded into the enclosure formed by the three-sided concrete wall. The quartz capsules were removed from the shipping containers and placed into a mechanism which crushed the quartz and allowed the lanthanum chloride to be dissolved in hydrochloric acid. The resulting solution was diluted with distilled water, neutralized with sodium hydroxide to approximately pH 2, and pumped to a holding bottle mounted on the side of a transit-mix concrete truck. The truck had previously been loaded with the soil and with the drum rotating, the radioisotope solution was sprayed onto the continually renewed surface of the tumbling soil. After the spraying procedure was completed, the tagged soil was transferred to a covered hopper on a dump truck. The hopper fed an adjustable spreader which was geared to the truck drive so that a constant mass of synthetic fallout was deposited on the surface over which the truck was driven. Slurry synthetic fallout was prepared by mixing dry synthetic fallout with water in a vehicle called a crash trailer. This trailer was equipped with a holding tank and a pump for spraying and recirculating.

The mass levels of synthetic fallout deposited were measured by placing foot-square pans on the areas before contamination. After the surfaces were contaminated, the pans were picked up and returned to the simulant processing building. Contents of each pan were weighed and the La^{140} content measured in a NRDL 4-pi ionization chamber. These measurements yielded mass deposited per unit area and average specific activity. An account of test procedures and decontamination results are reported in Reference 3.

During the operational phases of the tests, twenty batches of simulant were produced in twenty working days. Complete cooperation of the Air Force and the staff at the Material Testing Reactor made it possible to maintain a supply of radio-lanthanum for the tests.

Solubility measurements in the laboratory and in the field showed that the trivalent lanthanum was strongly adsorbed when sprayed on the Ambrose clay loam, and that desorption was negligible with the liquid decontamination methods used. However, no method was available for determining the specific activity of the tagged soil as a function of particle size in the sub-sieve range. Conventional dry sieving methods yielded sieve fractions, which when weighed and counted, gave distribution curves typified by those shown in Fig. 1. Inspection of the curves shows that a

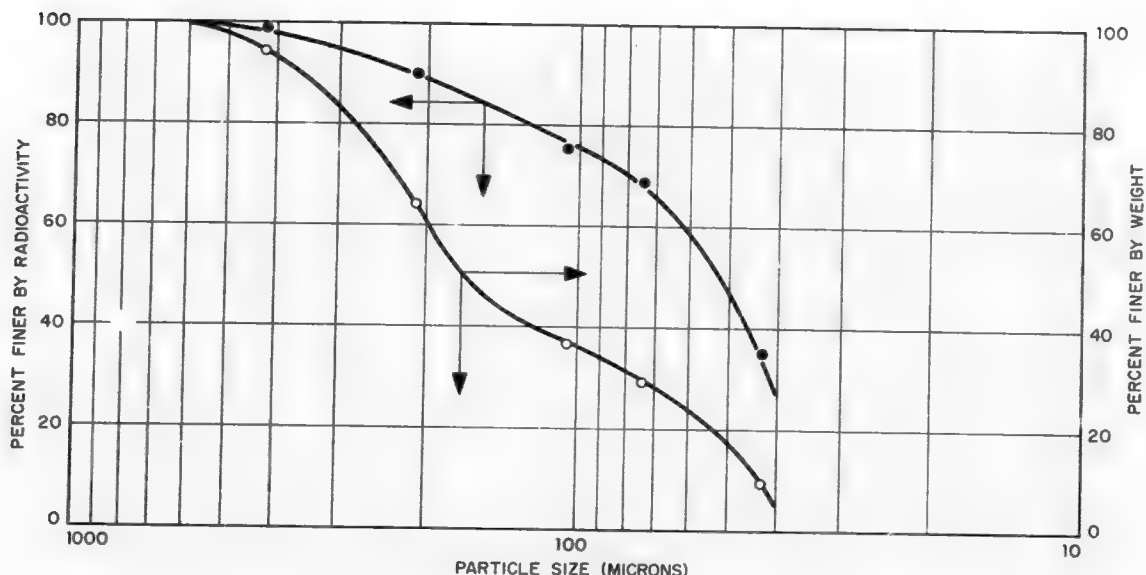


Fig. 1 Sieve Analysis of Stoneman I Synthetic Fallout Particles

large fraction of the lanthanum activity was associated with the small soil particles, and that 35 % of the original activity used was not accounted for by the analysis.

If one assumes that all particle sizes are decontaminated with equal efficiency by a decontamination method or, in other words, that fractionation of particle sizes does not occur in decontamination, then measurements of the gamma radiation before and after decontamination can be converted to weight units by the gross specific activity, and the decontamination effectiveness can be reported in terms of mass removed. No data were available at the time to verify such an assumption; in fact, it is unlikely that very small particles decontaminate the same as very large particles. Therefore, the production methods of synthetic fallout would have to be changed, if the decontamination results were to be reported in terms of mass, to produce a synthetic fallout in which each particle can be traced by an amount of activity proportional to its mass. In this case the curves in Fig. 1 would superimpose one upon the other and the selective removal of certain particle sizes by a decontamination method would then not affect the validity of converting gamma readings to mass units since the ratio between the two would be independent of particle size.

CHAPTER 2

PREPARATION OF SYNTHETIC FALLOUT FOR STONEMAN II

2.1 LABORATORY PHASE

Synthetic fallout for Stoneman II was limited to the simulation of fallout from land detonations, and again Ambrose clay loam was used as the bulk carrier and La^{140} as the radioactive tracer. However, as was previously noted, it was necessary to develop a method of tagging the soil particles so that a more reliable conversion factor between gamma radiation measurements and mass units could be determined. For this in turn, an analytical method for the determination of both mass distribution and activity distribution as a function of particle size (including the sub-sieve range) was a requisite.

2.1.1 Sub-Sieve Measurements by Air Sedimentation

Particle size may be defined in several ways, ranging from the actual particle dimensions to the diameter of an equivalent sphere. Thus large particles are usually sieved, and small particles may be measured under a microscope. However, the microscope method is tedious, the sample size is very small, and there is no apparent way to determine radioactive content of a particle by this method. Since terminal velocity is an important consideration in the distribution of fallout and in the determination of a contamination pattern, a method based on air sedimentation seemed promising for the determination of sub-sieve particle size distribution. In this case the particle diameter is related to an equivalent sphere and is determined by the terminal velocity of the particle for particles which obey Stokes' law. An apparatus using this principle was constructed, and preliminary evaluations were made of its utility for measuring the particle size distribution of both mass and activity.⁵ Figure 2 shows a schematic diagram of the apparatus

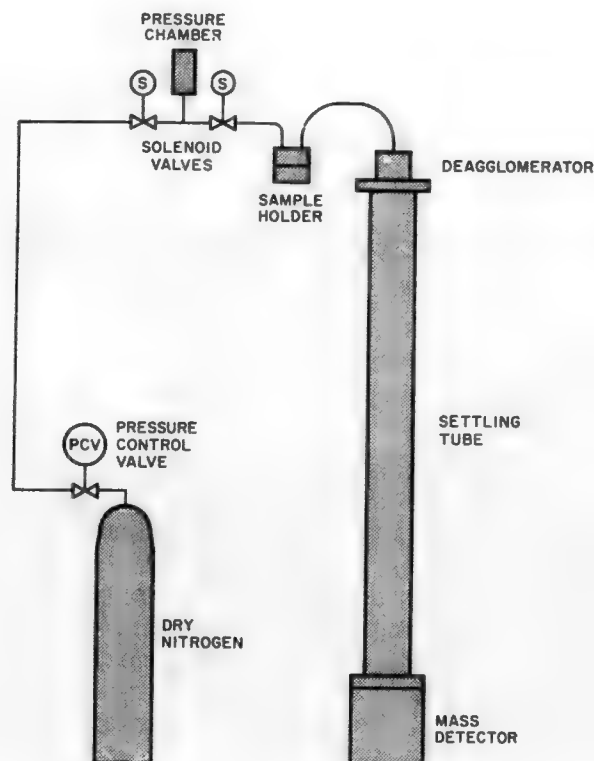


Fig. 2 Schematic of Air Sedimentation Column

which consisted of a deagglomerator unit at the top of a thermally insulated, vertically mounted tube with a radiation detector at the bottom.

The deagglomerator was similar to that of the Sharples Micromerograph,⁶ and was designed to reduce agglomerates to the basic unit particle. It consisted of an accurately machined and highly polished cone which fit a mating conical seat, and the spacing between the cone and seat was adjustable through a micrometer screw. A sample charging system consisted of two solenoid valves, a small pressure chamber, and a sample holder. The pressure chamber was filled with N_2 by opening the first solenoid valve. Opening the second valve discharged the N_2 through the sample holder into the deagglomerator. This turbulent flow of nitrogen picked up the solid particles from the holder, forced them through the deagglomerator and introduced them into the settling tube. The settling tube was constructed from 5 sections of glass pipe each four feet long and four inches in diameter. The tube was thermally insulated to prevent eddy currents which would occur from temperature differences along its surface. After the particles had

settled down a 20-ft tube they were collected on a Mylar plastic film at the bottom. Two different radiation detecting systems were used to record the time of arrival of the particles on the Mylar film; one was a sodium iodide crystal which recorded the gamma radiation from tagged particles as they arrived, and the other was a beta backscatter device which recorded the arrival of mass on the Mylar film. This required two separate tests, and the latter had to be conducted with soil which had not been tagged with a radioisotope.

Two possible limitations in the use of the apparatus were considered; however, neither had been previously investigated. It is possible that the charging burst of nitrogen gave the particles an initial velocity, and that the true starting time could not be accurately determined, and therefore particle diameters could not be determined by Stokes' law. A device was constructed which permitted the sampling of the particles which were arriving at the bottom of the tube at preselected times. Eight microscope cover glasses were mounted on a rotating stage which was covered by a shield. The shield had a 1/2-in. hole exposing the stage, and rotation of the stage exposed individual cover glasses for short intervals. The slides were examined under a microscope and the particle sizes plotted against the times of collection. An excellent fit was found with Stokes' law, showing that in this apparatus the effects of the charging bursts were insignificant. The other possible limitation was that particles were impinged on the tube and lost from the total sample when they adhered, and it was thought that the small particles would be most likely to exhibit this tendency. The particle size distribution of samples, each taken from the bulk sample, the residue in the holder, the walls of the tube, and the deposit on the Mylar film was determined with a microscope; the differences found were random and did not reflect particle size fractionation in any part of the apparatus.

Several runs were made with both the beta backscatter mass detector using inactive soil and with the gamma detector using radiotracers. Typical results are plotted in Fig. 3. While the apparatus had promising possibilities, it was abandoned as a practical method for field testing for several reasons; it was not portable, it was not convenient to measure both radioactive content and mass of a given particle size, and it required two separate runs on different samples to obtain the necessary data.

2.1.2 Sub-Sieve Measurements by Liquid Sedimentation

Another mechanical method of analysis was attempted, of which the fundamental basis again was Stokes' law, whereby the terminal

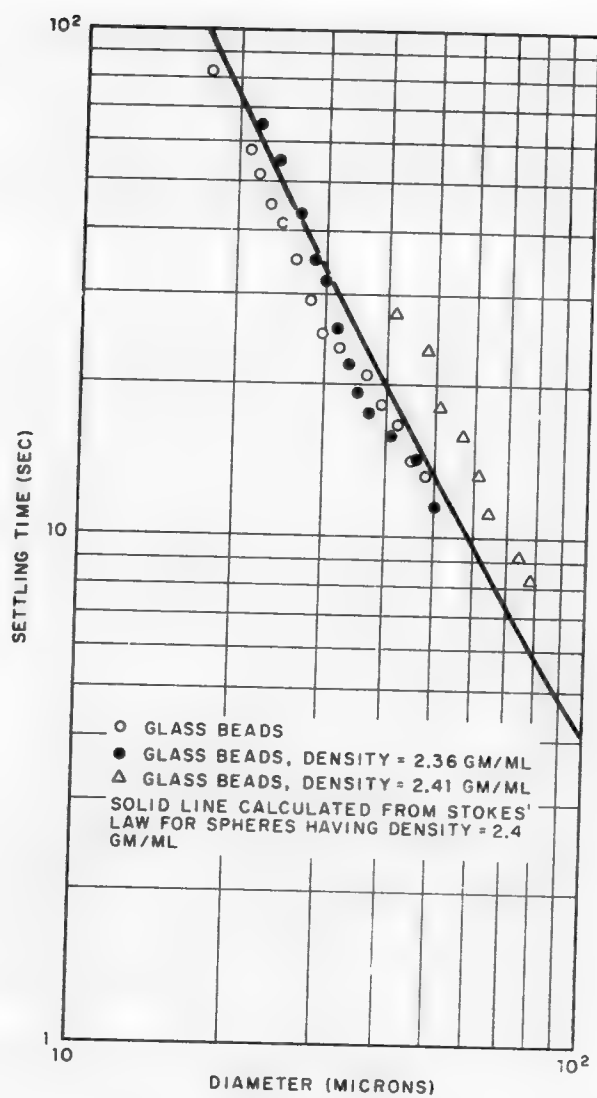


Fig. 3 Particle Size Distribution
by Air Sedimentation

velocity of the soil particles settling in a column of water was used to measure an equivalent diameter. While liquid sedimentation methods have certain classic objections such as dissolution of the particles in the liquid and a very short mean-free-path, the possibility of error through solubility of the tracer was known to be not critical. Several decontamination procedures utilize liquid methods that deagglomerate the particles in a manner similar to the sedimentation method, and the lanthanum ion was known to be highly adsorbed under all conditions encountered in liquid decontamination.

The somewhat unique requirement that both mass and activity be determined on the same sample placed limitations on a method which would be acceptable. Obviously, a hydrometer method which continually measured density would not measure the activity; however, the pipette method described in Ref. 7 seemed promising. In this method a sample of soil is suspended in water in a vertical column. At selected times, aliquots of the suspension are removed by pipetting from a fixed depth beneath the liquid level. Successive aliquots taken at increasing time intervals differ in that increasingly larger particles are missing from increasingly later samples, having settled past the sampling point. When the separate samples are weighted and counted the results give a mass distribution as a function of particle size and an activity distribution as a function of particle size. A particle sizing method combining wet sieving and liquid sedimentation was developed which gave distributions from 250 micron to 3 micron diameter, and the results of the method could be obtained in 4 hrs.

Laboratory batches of Stoneman I type synthetic fallout were prepared and analyzed by this method, and typical distributions are shown in Fig. 4. It will be noted that particles larger than 60 microns comprised 50 % of the total mass and were traced by 19 % of the activity; while particles smaller than 20 microns comprised 20 % of the mass and were traced by a disproportionate 50 % of the activity. A curve showing cumulative surface area is included to indicate that the radioisotope adsorption by the particles was dependent on surface area as well as particle diameter.

2.2 TAGGING BULK CARRIER

Experiments were conducted to determine methods for tagging the soil so that each particle would be traced by an amount of radioisotope proportional to its mass. As previously noted this was a requirement for the conversion of gamma radiation measurements to mass units.

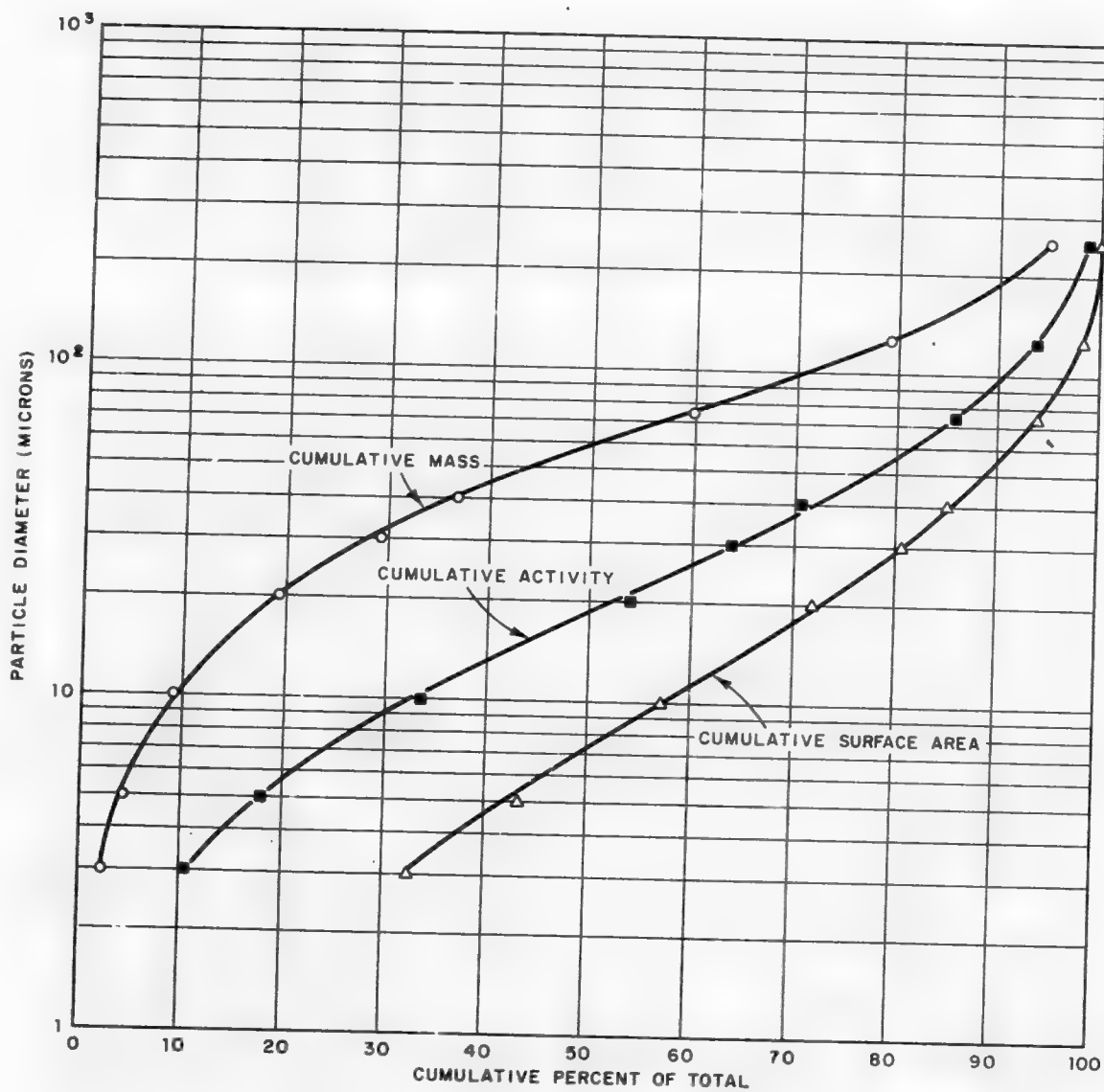


Fig. 4 Particle Size Distribution of (Stoneman I)
Synthetic Fallout by Liquid Sedimentation

Since the previous application of the tracer to the soil by spraying was essentially a dry process, it was postulated that this method did not represent equilibrium adsorption. Samples of the soil were immersed in solutions of La^{140} for periods from 1 hr to 4 days. No significant shift in the activity distribution in the direction of uniform specific activity with size was found.

In another experiment the soil was pretreated with inactive lanthanum in an attempt to saturate the adsorption properties of the small particles. However, when the radioactive La^{140} was added, the same high amount of adsorption on small particles was found.

The tagging of separate sieve fractions was next investigated. It was realized that this would increase the complexity of simulant production in the concrete mixer, and that the number of separately tagged fractions must be kept to a minimum. Also, since the fractions would have to be obtained by dry sieving multi-ton amounts of soil, the selected size ranges would have to be within the capabilities of commercial equipment. With these considerations in mind, the investigation was limited to a sieving operation which yielded plus 100 mesh, plus 200 mesh, and minus 200 mesh. These three fractions were separately tagged with La^{140} and the adsorption investigated as a function of increasing lanthanum concentration. It was found that while the small particles have greater adsorptive properties than large particles, the gross specific activity distribution with size appeared to be suitable. However, when the fractions were recombined and wetted, as would happen in the dispersal and liquid decontamination operations, the lanthanum exchanged from the larger particles to the small particles until an equilibrium similar to that shown in Fig. 4 was established. It was found that heating the soil to 500°C before tagging eliminated the exchange capacities since the organic materials were destroyed, and the lanthanum ions remained on the particles upon which they were placed.

2.3 BULK CARRIER

Fifty tons of Ambrose clay loam was obtained from the Camp Stoneman site and shipped to the Industrial Minerals and Chemical Co., Florin, California. The material was dried and crushed. Material larger than 60 mesh was eliminated, and all the remainder was heated in a rotary kiln to at least 500°C and then sieved into 3 fractions plus 100 mesh, plus 200 mesh, and minus 200 mesh.

2.4 RADIOISOTOPES

The Material Testing Reactor at Arco, Idaho was not available for La^{140} irradiations and it was found that the Los Alamos Scientific Laboratory, had the necessary quantities of La^{140} . Due to scheduling requirement at Los Alamos only one shipment per week could be furnished and calculations showed that 1000 curies of the La^{140} were required per week to insure 10 curies per day during the week's operation.

The Research Directorate, Air Force Special Weapons Center again offered to transport the shipments from Kirtland AFB to Travis AFB in C-47 aircraft.

CHAPTER 3

PRODUCTION OF SYNTHETIC FALLOUT FOR STONEMAN II

A dry synthetic fallout simulating fallout from a high-yield land surface burst was produced. The mass levels to be investigated were 10 grams/ft², 33 grams/ft² and 100 grams/ft²; which correspond to dose rates of approximately 300 r/hr, 1000 r/hr and 3000 r/hr all at one hour after burst. The areas to be contaminated required 500 lbs to 4000 lbs of synthetic fallout each day, and about 10 curies of La¹⁴⁰ per batch were required for adequate instrument response.

The synthetic fallout production consisted of the following: isotope procurement and delivery, processing of La¹⁴⁰, transit mix tagging of the Ambrose clay loam, dispersion of the material on test areas, and analytical studies. The flow diagram shown in Fig. 5 illustrates these steps.

3.1 ISOTOPE PROCUREMENT AND DELIVERY

The existing cavity in the LASL uranium shipping container limited the size and shape of the six stainless steel capsules. To facilitate handling, the capsules were placed in an aluminum basket as shown in Fig. 6. Also shown is the system of identification used to designate each capsule. Six sets of these capsules were constructed and shipped to LASL to insure that none would have to be reused during the operation.

Kilocurie separations of barium-lanthanum have been conducted at LASL for a number of years.⁸ In the procedure, the barium was precipitated as barium nitrate in 10 N nitric acid and filtered leaving the lanthanum in the acid solution. The La¹⁴⁰ content was determined by measuring its 2.15 mev gamma photon in a unique counter which only counts La¹⁴⁰ in the following manner.

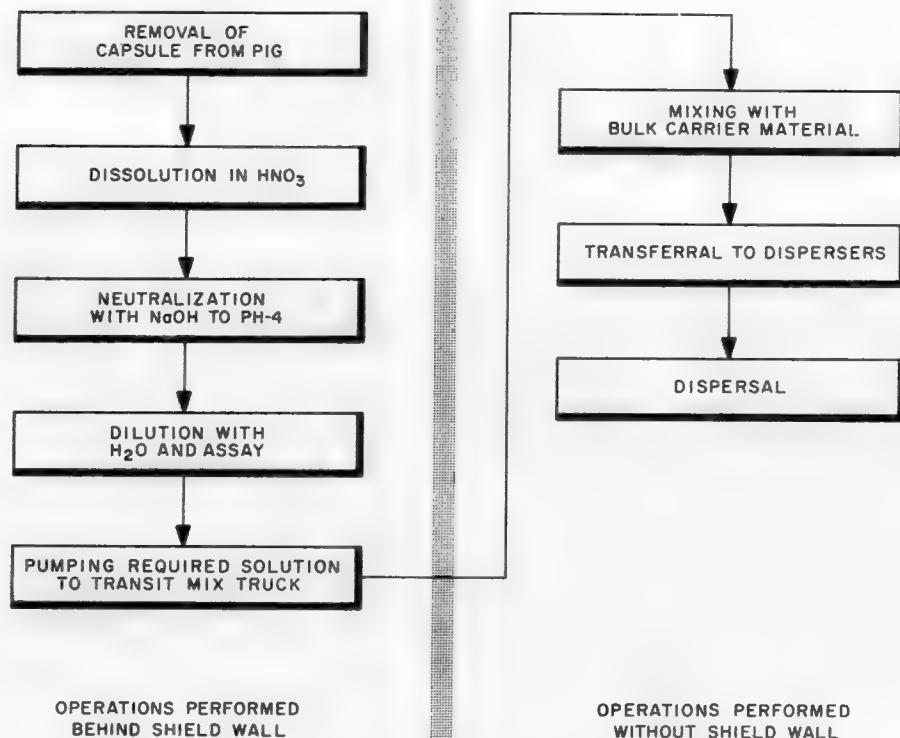
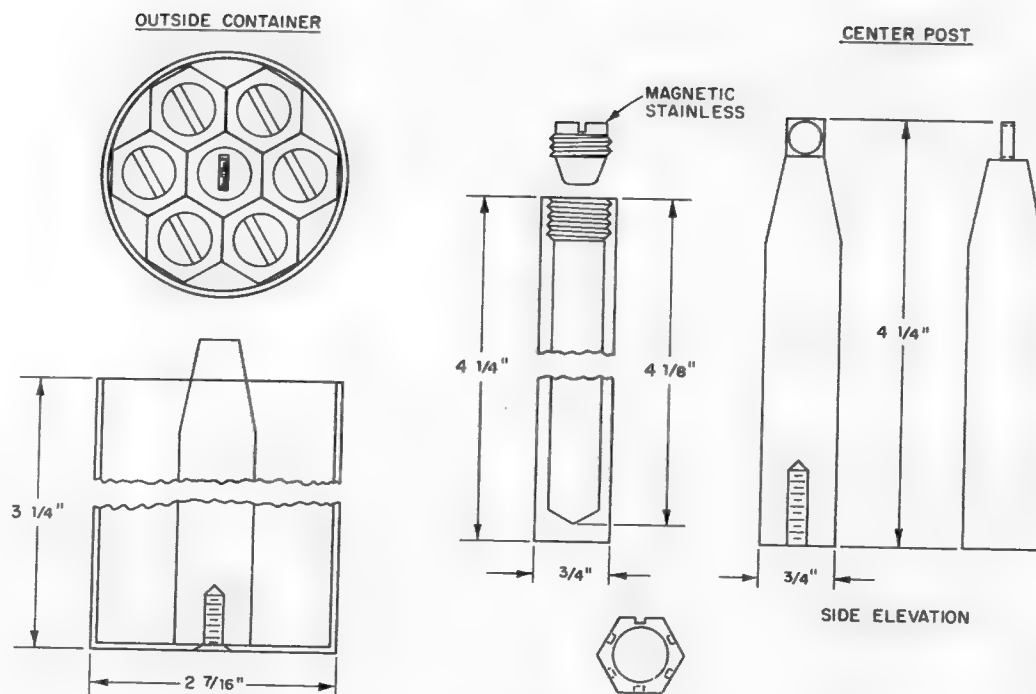


Fig. 5 Flow Diagram for Radioisotope Processing and Synthetic Fallout Manufacture



COLOR CODE FOR QUANTITY OF LA¹⁴⁰:

BROWN COLOR ON TOP OF ONE CHAMBER AND ONE NOTCH ON ONE SIDE 1/16 IN. WIDE AND 1/8 IN. DEEP

RED WITH ONE NOTCH ON TWO SIDES

ORANGE WITH ONE NOTCH ON THREE SIDES

YELLOW WITH ONE NOTCH ON FOUR SIDES

GREEN WITH ONE NOTCH ON FIVE SIDES

BLUE WITH ONE NOTCH ON SIX SIDES

Fig. 6 Sketch of La¹⁴⁰ Source Capsule and Basket

Gamma photons with energies greater than 1.63 mev on Be yield a neutron. The neutron on B^{10} yields an alpha and the alpha is counted on a standard crystal detector system. A volume of solution containing 1000 curies of La^{140} was removed from the cup and transferred to another section of the hot cell where the six capsules were filled and loaded into the uranium container. Table 1 indicates a typical shipment. After adding the aliquots to the capsules a heat lamp was used to evaporate the solution to complete dryness in order to make a spill enroute impossible.

TABLE 1
Typical Shipment of La^{140} Indicating Curie Contents

Capsule No.	Color Code	ml Added	Curies Added	Day of Use
1	brown	1.25	41.6	Monday
2	red	1.87	62.5	Tuesday
3	orange	2.96	98	Wednesday
4	yellow	4.41	147	Thursday
5	green	6.66	222	Friday
6	blue	<u>10.00</u>	<u>333</u>	Saturday
		27.15	904	

The separations were started on Thursday afternoon and completed Friday morning. A truck from Sandia Base transported the shipment to Kirtland Air Force Base and it was flown to Travis Air Force Base on Saturday or Sunday depending upon the availability of C-47 aircraft. The uranium container was picked up at Travis Air Force Base by Army truck and brought to Camp Stoneman. The basket containing the capsules was immediately transferred to another shielded container, and the uranium pig released to be returned on the same aircraft which brought it.

A NRDL representative accompanied the uranium container at all times, serving as a courier for all shipments and in a liaison capacity while at LASL.

Six almost identical shipments were made on successive weeks in fulfillment of the test requirements.

3.2 PROCESSING OF La^{140}

The concrete block shielding wall in Bldg. T-750 at Camp Stoneman was again used with a few modifications. A pair of Argonne Model 4 master-slave manipulators and a water-filled viewing window were provided to replace the Model 8's and the zinc bromide filled window, since these units were in service elsewhere. A 6000 lb lead pig was positioned behind the wall and accessible to the slaves to receive the basket full of capsules when they were transferred from the uranium container. The lid for this lead pig was made to rotate, and a series of indentations served to index a 1-in. hole over one capsule at a time. A magnet on the end of a flexible shaft was inserted into the hole to withdraw the correct capsule for the daily batch of synthetic fallout. The glass apparatus shown in Fig. 7 was mounted on a dolly which permitted the whole unit to be moved. Thus in the event of malfunction or a serious spill, the unit could be replaced without seriously delaying the operation. A shielded sump bottle was provided to receive radioactive solution which was excess to the daily needs. A complete description of the operation will be presented in Section 3.5.

3.3 TRANSIT MIX TRUCK AND BULK CARRIER

3.3.1 Transit Mix Truck

A 3-1/2 cu. yd. JAEGER Model 3HM mixer mounted on a Diamond T truck, Fig. 8, was modified to meet requirements for the production of synthetic fallout. A heavy walled glass pipe of 4 liter capacity was securely capped at both ends and provided with pipes and valves to permit filling from the top and discharging from the bottom either by gravity or by pressurizing. The assembly was rigidly mounted to the side of the truck, and reach rods were provided for operating the valves with a minimum of radiation exposure to personnel. The hollow shaft in the head of the drum which was normally used for adding water to the concrete mix, was used as an entry port for the contaminant spraying system. The contaminating solution was carried in 1/4-in. copper tubing through a flow meter and through the hollow shaft to an atomizing nozzle. Another 1/4-in. copper tube connecting to the nozzle through the hollow shaft was used to provide compressed air for atomizing and for keeping the solution jets clean. The nozzle was mounted on an arm extending upward from the hollow shaft, and a shield was placed over the assembly to protect the nozzles from falling soil. The concrete dumping chute was removed and replaced with an enclosed system which enabled quick transfer of the synthetic

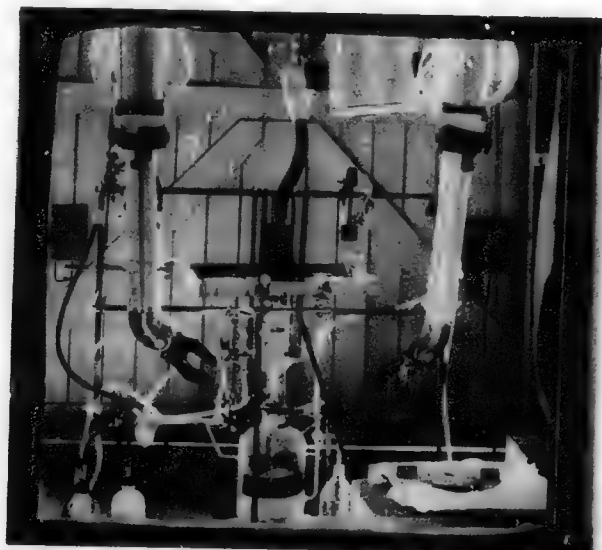


Fig. 7 Hot Cell Apparatus, View Through Shielding Window Showing Operation of Master Slave Manipulators and Glassware



Fig. 8 Transit Mix Truck Used for Mixing Synthetic Fallout

fallout with a minimum of spill and dust. The normal loading opening for concrete aggregate was covered and the inspection hatch on the drum was utilized for loading the dirt. A loading dock was constructed to facilitate soil handling and minimize radiation dosage to personnel. The existing water storage tank served as a shield for the driver.

3.3.2 Bulk Carrier Procurement

As previously noted the Ambrose clay loam was to be heated to at least 500°C in a rotary kiln and sieved into three size fractions. Constant liaison was maintained with the contractor, and it was soon obvious that his completion schedule would not be met. The first small shipment of processed soil arrived at Camp Stoneman on the day the operation was scheduled to start. A quality check on these fractions (by sieving on a Rotap) revealed that none were clean and that each contained large amounts (up to 20 %) of other sizes. Throughout the tests the procurement of soil proved to be the most unpredictable phase of the operation; the delivery schedules were always behind, and the quality of the sieving was never satisfactory. A NRDL representative was sent to the contractor's plant at Florin, Calif., mid-way in the series of tests, and his constant attention to the soil processing operation improved an otherwise unworkable situation.

3.4 FALLOUT DISPERSER AND TEST AREAS

3.4.1 Dispersers

The contaminant was dispersed over paved areas and land areas from a Burch Hydron Spreader (Fig. 9) (manufactured by the Burch Corp., Crestline, Ohio). The spreader was mounted on the rear of a 2-1/2 cu. yd. dump truck and was fed from an aluminum hopper which had a capacity of 2000 lbs. Raising the truck bed to the up position fed the soil to the Hydron spreader, and a positive displacement feed roll dispersed the contaminant in a 7 ft wide path. The feed roll was powered from the truck's drive shaft and could be operated by the driver from the cab. The quantity of soil dispersed was determined by the selection of one of several drive gears, the adjustment of the spreading lip, and the changing of sprockets in the linkage to the feed roll. The free fall drop of the soil was limited to about 2-in. to minimize the effect of wind disturbance.

The contaminant was dispersed over roof areas from a hand pulled garden spreader (Fig. 10) (O.M. Scott and Sons, Marysville,



Fig. 9 Disperser Truck Used for Dispersing Synthetic Fallout
Material on Land and Paved Areas



Fig. 10 Hand Spreaders for Dispersing Synthetic Fallout on Roofing Areas

Ohio, "Estate" Model 100). The hopper had a capacity of about 200 lbs of soil and a spreading width of 35 inches.

3.4.2 Test Areas

A map of Camp Stoneman indicating the various test areas is shown in Fig. 11. A description of the test areas utilized for each of the objectives follows:

Paved Areas

The asphalt test surfaces for the wet decontamination procedures were primarily located in area 5, a large paved parade ground. Each test area was 40 ft X 150 ft and to facilitate drainage and the recontamination of areas located down-slope, asphalt curbs were installed between each test area on the long axis. The asphalt surface was free of large cracks, holes or patches. The average slope of the areas was 1.5 %.

A limited amount of concrete test surfaces was available at the west end of Area 5. The surface was relatively free of cracks except for expansion joints which were filled with an asphaltic compound. Both dry and wet decontamination procedures were evaluated on these concrete test areas.

For the evaluation of the dry decontamination procedures, asphalt test areas were located on 21st and 22nd streets between Davis Avenue and Indiantown drive. These consisted of 20 ft X 100 ft test areas on asphalt-macadam crowned roadways.

Roofing Areas

The evaluation of wet decontamination procedures on roofing areas was limited to the two types of surfaces on existing buildings, tar and gravel and composition shingle. The tar and gravel had been exposed to the weather for eight years and the composition shingle for seven years. Table 2 gives descriptive details of the Roofing Surfaces.

Unpaved Areas

The four areas labelled No. 6 in Fig. 11 were utilized for the evaluation of the land reclamation procedures. Each of the four areas were prepared prior to the field test to represent four different surface conditions as follows:

(a) A moist surface with green grass. The land area immediately west of 14th Street and North of Davis Avenue was levelled,

TABLE 2

Descriptive Details of Roofing Surfaces

Test Surface	Material Description	Slope of Surface	Test Area Location
Tar and Gravel Roofing	5 plies tarred felt with a gravel finish	flat	Bldgs. T-602 T-603 T-604
Composition Shingles	Johns-Mansville Asphalt Strip Shingles	6 in./ft	Bldgs. T-1302 T-1313 T-1328 T-1329 T-1388 T-1379

tilled and planted with rye grass which by the start of the field test was typical of a large lawn area.

(b) A tilled moist surface. The land area north of Davis area and between 16th and 18th Street was leveled, tilled and kept in a moist condition.

(c) A dry tilled surface. The land area west of 16th street was leveled, tilled and kept in a dry condition.

(d) A dry land surface with withered vegetation. The land area at the south edge of the test site was not prepared except for the mowing of the existing vegetation to a height of six inches.

At each of the above locations, two 100 ft X 400 ft test areas were staked out.

3.5 ANALYTICAL FACILITIES AND MEASUREMENTS

3.5.1 Analytical Facilities

Laboratory facilities were provided by a mobile trailer parked in Bldg. T-745, and an existing air conditioned room in the same building was utilized as a balance room. Dark room facilities and a microscope were available in another building.

Camp Stoneman, California

PITTSBURG

Direction of Prevailing Winds during August-September

AREA A

1. HOT CELL
2. PROJECT H
3. CHANGE BO
4. RAD-SAFE
5. PERSONNEL
6. QM CORPS
7. PAVED TEST
8. UNPAVED T
9. LIVING AR
10. HQ, 11. SOFT
12. HESS HALL
13. a. SOIL M
14. b. TRANS
15. TRUCK
16. RADIOACTI
17. ENVIRONME
18. ENVIRONME

SCALE
0 1000
MILES IN FEET

Camp Stoneman, California

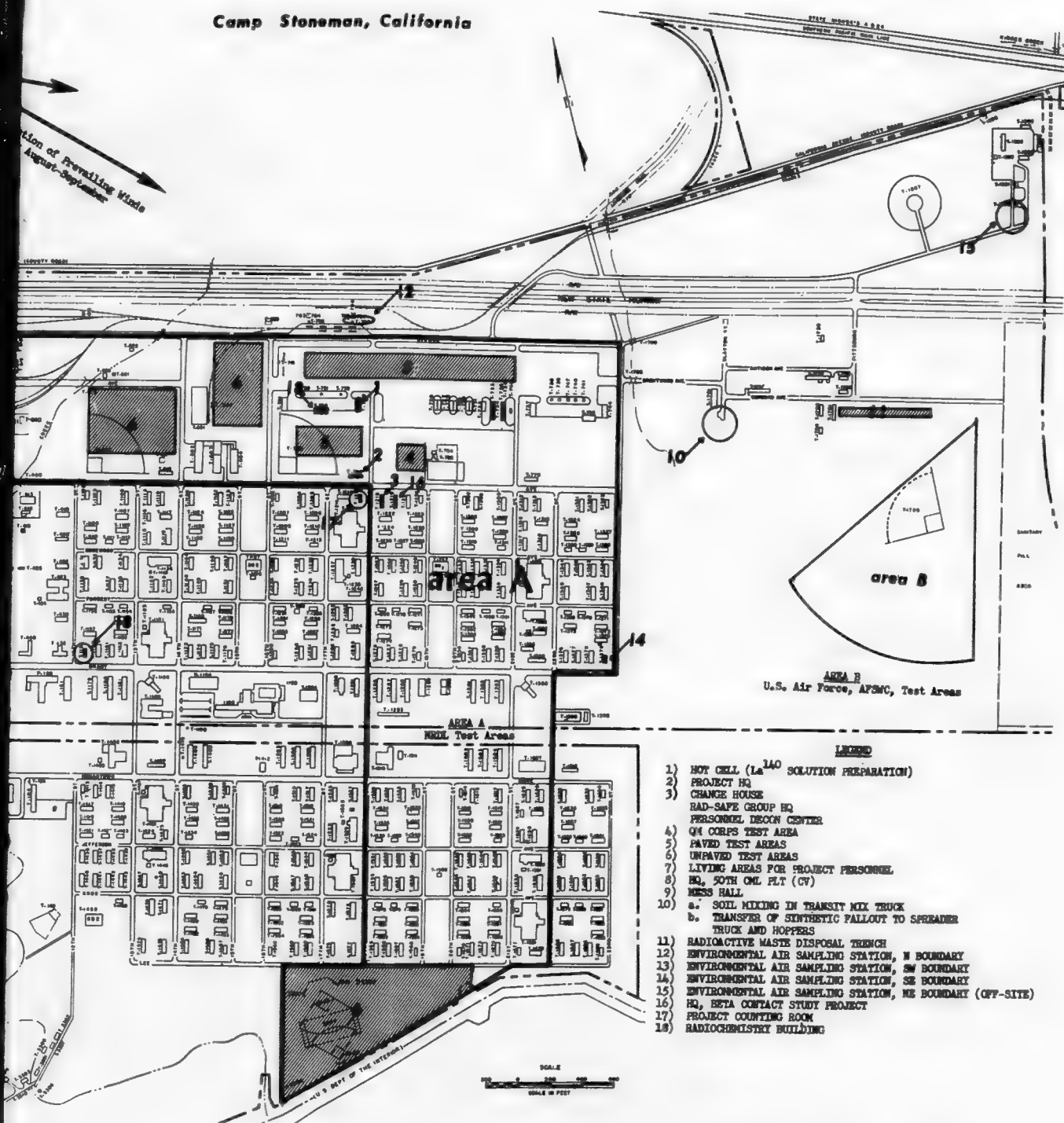


Fig. 11 Camp Stoneman, California

(B)

Counting facilities were located in Bldg. T-1215 about 1/4 mile from the hot cell to insure a reasonably low background. An NRDL 4-pi ionization chamber was used as the primary instrument and its milliamp response to La^{140} was related to d/s by,

$$d/s = 19.8 \times 10^{-14} \text{ ma}$$

when the readings were normalized to a standard chamber response of 560 ma for 100 micrograms of Ra^{90} . A low geometry scintillation crystal counter (Cat House) was provided to measure large samples and its response was related to the ionization chamber by the method shown in Section 3.4.3. A gamma spectra was obtained on each shipment of isotope with a single channel gamma analyzer to verify the radiochemical purity.

A mobile shielded gamma scintillation detector unit, Fig. 12, was used to determine the effectiveness of the various procedures evaluated by obtaining measurements of the radiation levels present on the test areas just prior to contamination (background), after contamination, and after decontamination. The detecting element of this unit consisted of a one inch NaI (Tl) Scintillation Crystal on a photomultiplier tube. The crystal and photomultiplier tube were mounted within a lead shield having a wall thickness of six inches. The shield is so mounted as to place the center of the detector one meter above ground plane. A collimated aperture subtending a solid angle of 50° permits entrance of radiation into the sensitive volume. Due to the geometry of this system approximately 98 % of the total radiation flux measured by the system from an ideal plane will fall within a circle having a radius of six feet. The entire unit is mounted on a four wheel trailer on which also was mounted a portable generator for the necessary power requirements and a housing enclosing the necessary electronic components of the system. A block diagram of the system is shown in Fig. 13.

The unit was towed over the test areas by a weapons carrier on which was attached an odometer that measured forward distance travelled in feet. This allowed the driver of the vehicle to position the unit at predetermined locations on the test areas for the necessary readings.

3.5.2 Analytical Measurements

Each shipment of processed soil was sieved and the results used to formulate daily batches of synthetic fallout. A complete analysis of each batch of synthetic fallout was conducted, and the results, including a sub-sieve distribution of mass and activity,



Fig. 12 Mobile Shielded Gamma Detector Unit

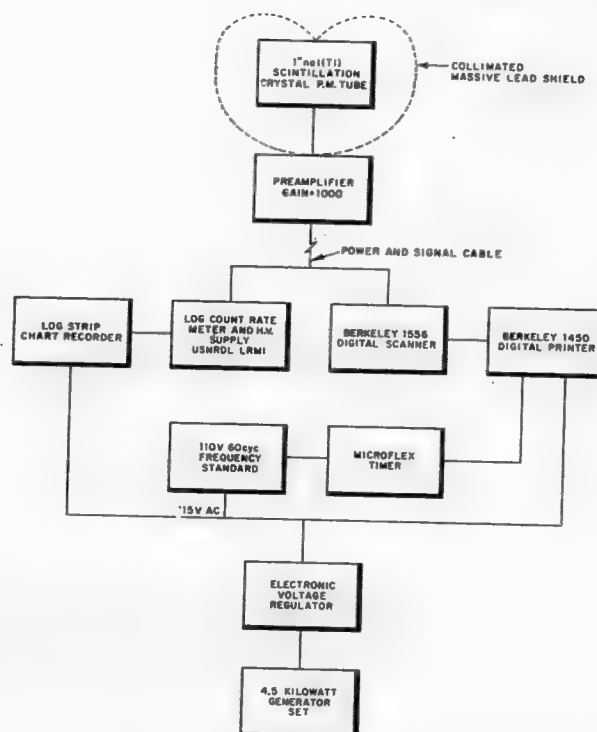


Fig. 13 Block Diagram of Egghead III Instrumentation

were available on the same day that the material was used. The mass deposited on each area was measured by collecting a sample in a 1.22 sq ft aluminum pan. The pan's contents were counted and weighed to give a deposited mass and a specific activity. Contaminated and decontaminated areas were monitored by the mobile shielded gamma scintillation unit.

3.5.3 Calibration of Mobile Shielded Gamma Scintillation Detector Initial Calibration

The Mobile Shielded Gamma Scintillation Detector, hereafter referred to as the Shielded Detector, was calibrated using three different techniques. The calibration factor designated K, relates relative activity (obtained from the shielded detector) to absolute activity (obtained as specific activity) for a unit area. The actual units are counts per disintegration per sq ft. For the initial calibration, Stoneman soil (Ambrose clay-loam), tagged with La^{140} , was distributed uniformly over a four foot diameter. Readings were taken at a height of 1.0 meter above the center of the circle. The results for three mass levels are shown below:

	Mass/area, g/ft ²	1.917	19.803	198.94
	Total activity, d/x	20.0 X 10 ⁴	208 X 10 ⁴	2091 X 10 ⁴
(1)	Activity/Area, d/s/ft ²	1.592 X 10 ⁴	16.57 X 10 ⁴	166.4 X 10 ⁴
(2)	Ave. Detector 'Rdg. c/s	8.6	89.5	858
	K, 2/1, c/d/ft ² X 10 ⁻⁴	5.404	5.403	5.157
	K, c/d/ft ² X 10 ⁻⁴	5.779	5.778	5.515

K was obtained by dividing K by .935 which represents that fraction of an infinite field which a 4-ft diameter circle presents to the shielded detector.

Daily Calibration

Prior to each day's run the shielded detector was calibrated with a sealed radium source. The source was placed on a jig at two different heights below the center of the crystal and a one minute count taken at each height. If the readings obtained, corrected for background, were out of line, necessary adjustments were made.

Calibration of Individual Tests

A calibration factor was calculated for each test to account for individual test variables such as surface type and initial mass loading. The data collected for this purpose include: (1) weight of the material dispersed as measured by collectors (1.22 sq ft shallow pans) placed approximately every 400 sq ft; (2) the gross activity

of the pan sample measured in a large sample counter (cat house); (3) the specific activity of a 5 gram sample taken from the pan and measured in the USNRDL 4-pi ionization chamber; (4) the average of the readings taken with the shielded detector on the contaminated area. The relationship of these items is as follows:

Let

- c_1/s = gross intensity, in cps, of 1.22 ft² pan sample (cat house)
- $I_r - c_2/s$ = initial intensity of contaminated surfaces in cps (mobile shielded detector)
- $R_r - c_3/s$ = intensity of decontaminated surface, in cps (mobile shielded detector)
- S = specific activity in disintegrations per second per gram (d/s)/g (4-pi ion chamber)*
- g_1 = weight of contaminant per 1.22 ft² pan, grams
- K = calibration factor, c(ft²)/d
- M_o = calculated initial mass level, g/ft²
- M = calculated final mass level, g/ft²
- C = a mass conversion factor, (c/s)/(g/ft²)

*The calibration factor used, taken from USNRDL-TR-155 (Ref. 9), was

$$3.30 \times 10^{-15} \frac{\text{ma}}{\text{d/m}}$$

- (1) The relationship between the gross activity of the sample and the specific activity is

$$\frac{c_1/s}{\frac{d/s}{g}} = c_1/d$$

The ratio, c_1/d , should be a constant for all cases.

- (2) Further

$$\frac{c_1/s}{c_1/d} = d/s$$

- (3) Entering an area term, (2) becomes = $\frac{d/s}{\text{ft}^2}$

- (4) Obtaining the relationship of the initial intensity to (3) -

$$\frac{\frac{c_2/s}{d/s}}{ft^2} = \frac{c_2(ft^2)}{d} = K$$

K is theoretically a constant for identical surfaces and mass levels.

- (5) A conversion factor can be derived by multiplying K by specific activity.

$$K \times S = \frac{c(ft^2)}{d} \times \frac{d/s}{g} = \frac{c_2/s}{g/ft^2} = C$$

Thus

$$(6) \quad \frac{I_r}{C} = \frac{\frac{c_2/s}{g/ft^2}}{g/ft^2} = M_o (g/ft^2)$$

In the ideal case $M_o = g/ft^2$

Assuming specific activity, S, and K are both constant for all measurements taken during the course of the test, the residual intensity data can be treated in the same manner.

$$(7) \quad \frac{R_r}{C} = \frac{\frac{c_3/s}{g/ft^2}}{g/ft^2} = M(g/ft^2)$$

3.6 ACCOUNT OF THE PREPARATION AND DISPERSAL OF A BATCH OF SYNTHETIC FALLOUT

Batches of synthetic fallout were designated by the capsule which was used to radiotag them. Thus, 4.5 was from the 4th week's shipment and Friday's capsule. The history of one batch, 4.5, of synthetic fallout followed will be to demonstrate the work done in the twenty cycles required for the Operation. The contamination of 8,000 ft² of paved and roofing areas with 1 ton of synthetic fallout was scheduled for Saturday's operation (the day the 4.5 batch would be used).

On the morning of 11 September 1958, the two men assigned to the hot cell operation dressed out in coveralls, booties, rubber

gloves and surgeon's caps. The radiation levels on the slave side of the shielding wall were checked with a TLB to insure working safety. The fritted filter was cleaned, stopcocks were inspected and lubricated, and tygon tubing lines were checked for pressure and vacuum tightness. The required volumes of nitric acid and sodium hydroxide were placed on the working tray, and clean pipettes and volumetric glassware were provided. Special tools were located in their proper positions and the electrical circuits for magnetic stirrer, heat lamp and cell lighting were checked. A 10 ml test tube was securely fastened to the end of a long pole. The test tube was clamped in position under a side arm of the 4 liter flask so that a very small aliquot of solution could be delivered into it for assay. The other end of the long pole extended over the shielding wall to enable the sample to be removed from the cell. Surgeon's gloves were then removed before going to the master side of the shielding wall.

Checks were made to insure that no one was inside the 100 mr/hr line (designated by ropes and signs), and personnel operating in the test areas warned that the capsule would be withdrawn, (this was quite necessary since radiation from the capsule would contribute to effectiveness measurements on the test areas). The hot cell operations described from this point on were conducted with master-slave manipulators, and all personnel were protected by the shielding wall.

The rotating lid was indexed over the Friday capsule and the magnet inserted in the hole, as the capsule was lifted out, its identity was checked (5 notches and green). A TLB reading was used to insure that the activity was 10 curies within a factor of 2. The hexagon capsule was wedged in the holding jig, and the lid removed with a magnetic screwdriver. A pipette was used to deliver 10 ml nitric acid to the capsule and a heat lamp was used to warm the contents. The dry lanthanum nitrate was completely dissolved in 15 min., and the capsule was carefully inverted over a fixture which allowed the contents to drain into a funnel and onto a fritted filter. The fixture also allowed the inside of the capsule to be rinsed with distilled water. A vacuum was applied to the system and the solution transferred through the filter and into the 4 liter flask below. Rinsing was continued until the radioactive solution was diluted to 1 liter, and a magnetic stirrer provided vigorous stirring in the 4 liter flask. Fifteen ml of 6 N sodium hydroxide was added to adjust the pH to 2-4; this held the lanthanum in solution, and minimized attack on the metal tubing and nozzle. About 0.5 ml of the radioactive solution was drained into the 10 ml test tube, and removed from the cell on the end of the long pole. A 50 microliter aliquot of the solution gave 570×10^{-8} ma on the 4-pi ionization chamber, and

$$\frac{570 \times 10^{-8} \text{ ma}}{19.8 \times 10^{-14} \text{ ma}} \times \frac{1000 \text{ ml}}{0.05 \text{ ml}} \times \frac{560 \text{ ma}}{670 \text{ ma}} \times \frac{1}{3.7 \times 10^{10}} = 12.8 \text{ curies}$$

All of the solution was required for batch 4.5 of synthetic fallout, and at 1300 hrs the radioactive La^{140} solution was ready for delivery to the transit mix trucks.

The transit mix operations also started at 0700 hrs on 11 Sept. 1958 and the crew consisted of 3 men who were rotated on the various jobs to distribute the radiation dosage. Prior to the mixing operation, the truck was driven to a remote area of the base where waste disposal pits had been provided. The drum was thoroughly emptied and the solution spraying system was washed to reduce the radiation levels. The truck was returned to the garage where it was given a thorough daily maintenance check, and where the solution spraying system was inspected and tested.

The daily sieve analysis (by the analytical group) showed that on 11 September 1958 the composition of the soil fractions on hand was

	<u>plus 100</u>	<u>plus 200</u>	<u>minus 200</u>
plus 100 (red bags)	85 %	14 %	1 %
plus 200 (yellow bags)	10 %	69 %	21 %
minus 200 (green bags)			100 %

Calculations (described in results and discussion) of capture efficiency on the previous batch showed that the composition of the 500 lb mix for contamination should consist of

plus 100 (red bags)	324 lb
plus 200 (yellow bags)	145 lb
minus 200 (green bags)	41 lb

These quantities were carefully weighed ($\pm 1\%$) and placed on a fork lift which in turn lifted them to a hopper on the loading dock. The truck was backed into place beneath the dock and the drum rotated until the inspection plate was in an up position. The hatch cover was removed and the soil transferred from the hopper through a cloth sleeve to the drum. The hatch cover was securely lugged down and a man drove the truck into position alongside the hot cell building. The 1/4-in. tygon delivery tube leading from the 4 liter flask was coupled to the solution bottle on the truck. Pressure was applied to the 4 liter flask in the hot cell and the 1000 ml of radioactive solution was transferred to the bottle on the truck. A 500 ml water wash was pumped through the system and the delivery tube disconnected.

The truck was driven to an area where the spraying could be conducted without interfering with other operations. The atomizing air was turned on and set, at 10 lbs, the rotating drum was started, and the solution flow was adjusted to 40 cc per minute. A transit was focused on the flow meter from about 50 yards and the operation observed from this point. The radioactive solution was sprayed in 37 min. and was followed by a 500 ml wash. Mixing was continued for 30 min., after which the contaminated soil was sampled. The sample was sieved and when the fractions were counted and weighed the following mass activity relationship existed.

	<u>Mass</u>	<u>Activity</u>
plus 100 mesh	62.9 %	31.6 %
plus 200 mesh	22.2 %	21.8 %
minus 200 mesh	14.3 %	46.6 %

Also an assay of 10 grams of the 500 lbs of contaminated soil showed 570×10^{-8} ma

$$\frac{570 \times 10^{-8}}{19.8 \times 10^{-14}} \times \frac{500 \text{ lb} \times 454}{10 \text{ gram}} \times \frac{560}{670} \times \frac{1}{3.7 \times 10^{10}} = 12.7 \text{ curies}$$

This indicated that all the radioactive solution had been retained on the soil (decay corrections from 1100 hours to 1415 hours were unnecessary). Again calculations were made (described in Chapter 4) to determine the quantities of soil fractions (non-contaminated) which would be added to the mixer to yield 2000 lbs. Thus the following amounts of soil were added.

plus 100 mesh (red bags)	187 lbs
plus 200 mesh (yellow bags)	500 lbs
minus 200 mesh (green bags)	819 lbs

These quantities were weighed, transferred to the loading dock, and added to the transit mix truck. An additional 10 minutes of mixing then thoroughly blended batch 4.5 of synthetic fallout. The truck was then driven to an area selected for transfer of the synthetic fallout material into the dispersers.

On the morning of 12 September 1958 the paved area dispersal crew consisting of two men, drove the dump truck used for paved area dispersal to the area where the transit mix was parked. By positioning the hopper on the rear of the dump truck under the discharge chute of the transit mix, a transfer of the synthetic fallout material from the transit mix to the dump truck was accomplished. The dump truck was then driven to the paved test area selected

for contamination and the synthetic fallout material was dispersed over the area at the amount previously selected, 100 grams/ft².

A hopper which held approximately 500 pounds of soil was used to transport the synthetic fallout material required for the contamination of roofing areas. The transfer from the transit mix truck to the hopper was made in the same manner as the transfer to the dump truck. The hopper was moved to the building on which the roofing test areas were located and the hand spreaders used to disperse the synthetic fallout material on the roofing areas were loaded through a canvas chute at the bottom of the hopper. A large fork lift was used to hoist the hand spreaders up to the roof levels. The dispersal crew for the contamination of the roofing areas consisted of four men.

The laboratory type analytical measurements were assigned to 4 men. Two men were charged with taking and measuring samples of the deposited mass, and they placed out and recovered a total of 32 aluminum pans from the 4 areas which were contaminated on 12 September 1958. The pans and their contents were placed in polyethylene bags and taken to the instrument building for counting, in the "Cat House". The pans were then transferred to the balance room and the synthetic fallout weighed. A 5 gram sample of soil from each pan was placed in a test tube and taken back to the counting room for measurement in the 4-pi ionization chamber. The measurements of distribution of mass and activity as a function of particle size were conducted by two additional men. On 12 September 1958 they received the sample of synthetic fallout from batch 4.5. Twenty grams of this sample were wet sieved on a 200 mesh sieve; the fraction remaining on the sieve (plus 200 mesh) was dried and resieved through a nest containing 60 mesh, 115 mesh, and 200 mesh, and the slurry fraction passing 200 mesh was transferred to a settling column. The glass column (3-in. i.d.) had a capacity of 2 liters and was vacuum jacketed to prevent eddy currents. A side arm with a stop cock was located 10 cm down from the 2 liter liquid level mark, and permitted the taking of samples from this depth and midway across a radius of the column. The slurry fraction was diluted to exactly 2 liters and vigorously shaken for 2 min. The column was then set upright in a perfectly vertical position and a timer started. Five aliquots were withdrawn into previously weighed 10 ml volumetric flasks at 61 secs, 104 secs, 221 secs, 13.9 min, and 52.3 min and 2.26 hrs, for cuts at 40 μ , 30 μ , 20 μ , 10 μ , 5 μ and 3 μ . The flasks were centrifuged for 10 minutes and most of the clear water decanted off, after which they were placed in an oven and thoroughly dried. The flasks were then cooled to room temperature, weighed, and measured in the 4-pi ionization chamber. The data sheet is shown in Table 3 and resulting curves are plotted in Fig. 14.

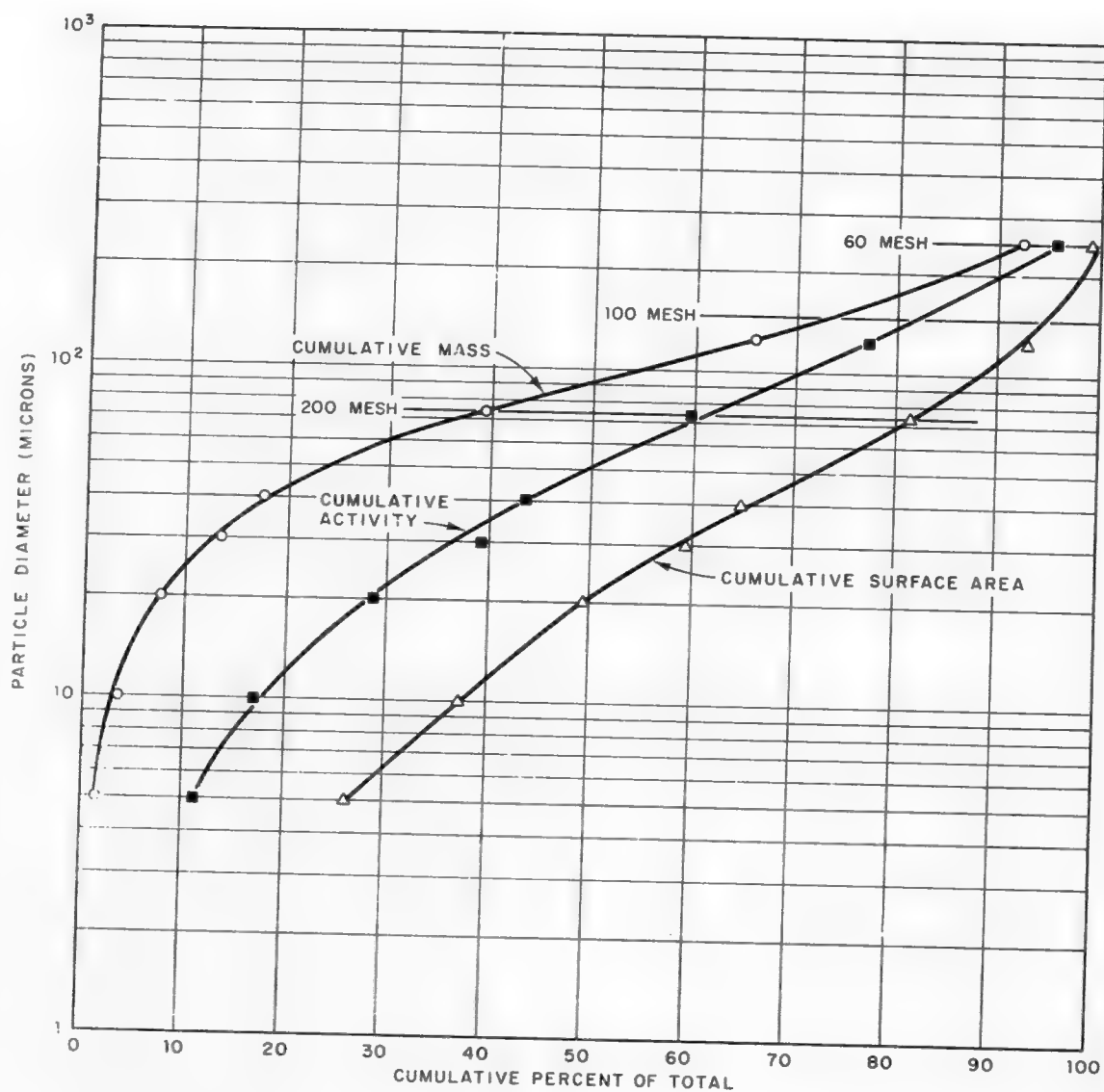


Fig. 14 Analysis of Batch 4.5 of Synthetic Fallout Material

TABLE 3

Capsule No. 4.5C, Week of 9/8/58

Particle Dia. (μ)	Cum. Mass (%)	Cum. Activity (%)	Cum. Surface Area (%)
250	92.4	95.8	99.1
125	65.9	77.1	93.0
74	39.3	59.6	81.3
40	17.7	43.5	64.8
30	13.3	39.1	59.3
20	7.6	28.7	49.4
10	3.4	17.0	37.2

CHAPTER 4

RESULTS AND DISCUSSION, AND CONCLUSIONS

4.1 RESULTS AND DISCUSSION

Decontamination studies have progressed in quite an orderly manner, though the progress sometimes appeared disorganized and even accidental. Countermeasure experiments conducted at field testing of nuclear devices have always been fraught with difficulty, primarily because the testing of weapons was the prime objective, and the countermeasure experiments had a much lower priority. Few of the countermeasure programs were able to accomplish all objectives, and many experiments were complete "failures". However, in most cases some data were obtained, and these were check points for the development of the theory of decontamination.² The use of radioactive synthetic fallout to study countermeasures on large areas has also developed by "cut and try" methods. Some experiments, as previously noted, yielded data that could not be interpreted; nevertheless, much was learned about the handling of isotopes, the tagging of carrier material, decontamination methods, and analytical measurements. Concurrently the theoretical studies gave new concepts of contaminating events (the chemical and physical properties of fallout and mass levels), explained significant factors in decontamination effectiveness (dose rate and dose), and pointed out data that were needed to further the studies. Thus by recognizing shortcomings of previous experiments and correcting them, and by utilizing available theory, systematic progress has been made in a science that was unknown but a few years ago.

The isotope procurement and the isotope solution preparation phases of the tests were so successful as to leave little for discussion. The success is best evidenced by the results: there were 10 curies of La^{140} solution for each day's operation; there were no incidents of malfunction or spills; no one received a radiation dose greater than maximum permissible levels.

The preparation of synthetic fallout in the transit mix truck was also completed on schedule, although several changes in plan were necessary. The non-availability of soil forced many substitutions. Batches 1.1 and 1.2 were made by the Stoneman I process, and Batch 1.3 was composed of excess harbor bottom material. When the sieve fractions were available they were not clean cuts; actually, they were not uniform from bag to bag. This criticism of the sieving is not meant as a reflection on the contractor, who was quite sincere; it is rather aimed at the difficulty of the task itself, and to demonstrate the difficulty in extrapolating laboratory experiments with grams, to commercial operations with ton quantities. The Ambrose clay loam was probably quite uniform after drying, crushing and removing the large material, but each successive operation made it more non-uniform. Heating in the gas-fired rotary kiln caused some particles to agglomerate by fusing or sintering, and the combustion gases carried off quantities of fines. The material was delivered from the kiln in surges, and the material in the surges was alternately large particles and small particles, thus the barreled product was not uniform. Separation into fractions was to be accomplished in one pass over a "Sweco" (a well known commercial screening machine) with a 100 mesh screen, and one pass over a sifter with a 200 mesh screen. The actual operation required 3 to 4 passes over the "Sweco" for the first cut, and when the sifter was found completely unsatisfactory it was necessary to change screens on the "Sweco" and make an additional 3 to 4 passes over a 200 mesh screen for the second and third fractions. An air elutriation machine was constructed but it was also unsuccessful in making the separations. Fired Ambrose clay loam is alleged to have unusual sieving properties, including electrostatic charges, incipient fusion, and screen blinding. None of these properties has been quantitated, but it suffices to say the soil is difficult to sieve. However, by utilizing whatever soil was available on a day-to-day basis, twenty batches of synthetic fallout were prepared.

The particle size distribution of mass and of activity was plotted from the data presented in Table 4. An inspection of the table shows that the ideal tagging (mass curve and activity curve superimpose) was never achieved, but improvement will be noted from one batch to the next. The data from batch 4.5 and 5.6 are plotted in Fig. 14 and Fig. 15 respectively. A comparison of these curves with those in Fig. 4 (Stoneman I) show the improved characteristics of the Stoneman II synthetic fallout.

For batch 4.5 it was desired to produce 2000 lbs of synthetic fallout having the following analysis:

Table 4

ANALYSIS OF SYNTHETIC FALLOUT

Capsule No.	Percent of Mass and Activity for Particles Less Than Indicated Size															
	250 μ		125 μ		74 μ		40 μ		30 μ		20 μ		10 μ		5 μ	
	M	A	M	A	M	A	M	A	M	A	M	A	M	A	M	A
1.1	95.1	99.1	79.4	95.3	58.3	88.5	30.2	78.3	23.0	74.1	17.3	69.9	7.6	41.9	3.8	29.3
1.2	94.0	98.1	76.0	90.2	53.1	80.2	28.5	72.4	21.5	66.0	14.0	61.5	6.6	43.4	3.4	27.9
1.3	92.8	98.4	70.0	90.1	48.4	78.8	23.4	64.2	18.1	61.3	11.6	53.3	5.5	39.3	2.9	26.1
1.4							62.1	92.0	42.8	84.0	26.7	70.6	11.2	43.5	5.2	26.0
1.5	94.1	99.8	80.3	99.4	70.1	99.8	49.9	95.5	41.5	94.0	31.2	88.5	17.9	71.2	12.8	51.5
1.6	88.0	99.0	53.7	92.9	29.3	85.9	15.8	81.0	11.9	75.9	9.6	73.1	5.2	54.0	2.3	33.8
2.1	90.6	95.0	73.2	81.1	62.9	74.0	41.0	55.4	39.3	53.3	25.3	44.5	10.1	27.2	4.1	15.7
2.2	94.2	92.9	73.2	57.7	48.1	30.7	23.2	24.5	17.2	24.8	10.9	18.5	4.5	11.4	1.9	8.3
2.3	95.0	95.5	73.9	67.3	34.0	42.8	11.0	34.6	7.3	30.5	4.2	24.5	1.9	16.5	1.4	11.6
2.4	95.6	95.5	63.2	68.2	41.4	46.7	13.1	32.8	8.6	27.4	5.5	22.6	2.9	17.1	1.5	12.6
2.5	90.8	96.8	62.7	73.2	46.5	56.5	15.3	39.2	9.2	34.5	5.0	27.4	2.2	21.1	1.2	15.5
2.6	95.2	97.5	84.2	88.1	43.3	50.5	12.1	35.7	7.8	33.4	4.3	27.4	1.9	19.4	1.0	15.1
3.1	95.4	98.6	85.4	93.2	57.6	74.0	23.8	58.7	16.1	54.5	9.9	46.1	4.9	32.7	2.1	19.6
3.2	95.4	98.7	80.4	92.8	51.9	77.9	23.0	60.2	15.4	55.3	8.8	42.4	3.6	23.6	1.7	15.8
3.3	95.1	97.5	77.5	84.3	56.1	70.2	24.4	56.6	16.6	48.8	9.1	35.7	4.1	21.1	1.8	12.3
3.4	95.7	97.8	81.7	86.3	65.8	71.5	26.4	57.1	16.6	50.5	8.7	39.4	3.9	25.5	2.0	15.6
3.5	93.3	96.8	71.1	78.0	46.6	61.5	18.5	47.4	13.6	40.8	8.0	31.8	4.1	20.0	1.9	12.0
3.6	93.3	96.4	68.5	80.0	39.1	60.7	21.0	48.6	15.1	43.4	9.4	37.6	3.4	35.7	1.1	7.4
4.1	95.0	95.9	71.3	77.1	39.3	56.8	21.6	50.7	15.8	47.3	8.3	36.3	2.9	23.2	0.9	14.1
4.2	94.5	97.5	80.8	88.1	47.7	60.1	19.5	47.4	13.2	42.2	7.8	33.6	3.6	21.3	1.6	14.0
4.3	93.5	96.7	69.6	80.5	43.4	63.8	20.5	49.6	14.8	42.4	10.0	33.5	4.4	22.6	1.8	14.0
4.4	91.6	95.1	62.1	72.2	25.8	39.3	10.2	22.6	6.5	19.0	3.8	14.9	2.0	9.9	0.8	5.4
4.5	92.4	95.8	65.9	77.1	39.3	59.6	17.7	43.5	13.3	39.1	7.6	28.7	3.4	17.0	1.5	11.2
4.6	92.8	93.9	70.4	75.1	42.4	57.8	21.3	41.2	14.8	35.9	8.2	29.3	3.4	20.1	3.0	13.7
5.1	87.1	91.4	59.9	71.8	46.7	56.8	25.9	46.0	19.5	41.5	11.9	32.7	4.0	17.9	1.5	10.9
5.2	89.3	92.1	58.1	65.0	47.4	52.4	23.2	47.4	19.2	45.7	11.7	35.4	4.2	20.9	1.3	13.3
5.3	89.8	90.0	58.7	61.1	48.1	46.0	27.9	39.0	20.9	34.2	12.3	26.1	4.8	14.9	1.5	8.0
5.4	89.6	88.7	59.4	61.6	49.1	51.5	23.8	39.3	9.7	28.6	5.3	23.3	2.2	16.3	1.2	11.3
5.5	92.5	90.7	66.3	62.0	50.0	46.6	23.5	33.8	17.7	31.8	9.1	24.3	3.2	14.8	1.1	9.0
5.6	95.4	95.1	75.5	70.7	50.1	48.3	29.5	34.4	21.8	29.0	13.6	21.8	6.0	13.0	1.5	6.5

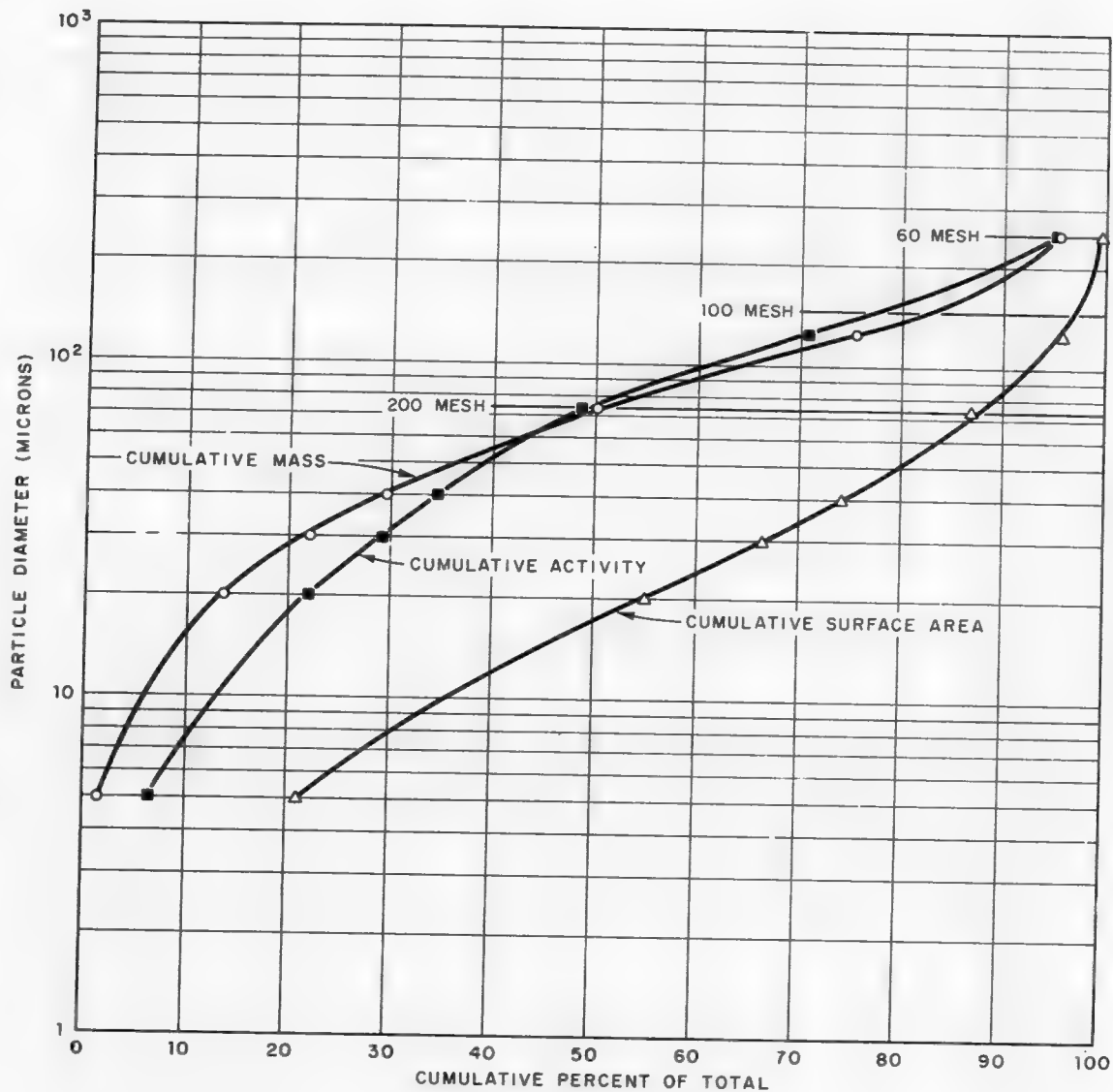


Fig. 15 Analysis of Batch 5.6 of Synthetic Fallout Material

plus 100 mesh	25 %
plus 200 mesh	25 %
minus 200 mesh	50 %

Capture efficiencies for the soil fractions were calculated from the sieving results from batch 4.4. An analysis of the original 500 lbs of contaminated soil gave

	<u>Mass</u>	<u>Activity</u>
plus 100 mesh	56.4 %	34.4 %
plus 200 mesh	32.0 %	37.3 %
minus 200 mesh	11.0 %	28.3 %

and after adding the inert soil and blending the final results were

	<u>Mass</u>	<u>Activity</u>
plus 100 mesh	36.5 %	26.4 %
plus 200 mesh	37.5 %	33.0 %
minus 200 mesh	26.1 %	40.6 %

For the computation of capture values the mass values of the original 500 lbs. must be used; however, the final values of activity distribution can be used, since adding inert soil could not have affected the distribution which existed.

	<u>Plus 100 mesh</u>	<u>Plus 200 mesh</u>	<u>Minus 200 mesh</u>
(1) % mass	56.7	32.2	11.0
(2) % activity	26.4	33.0	40.6
(3) capture, a activity/mass	.466	1.03	3.67
(4) normalize	1.00	2.22	7.78
(5) % desired	25	25	50
(6) weight, %/ capture	25	11.7	6.45
(7) normalize to 100 %	57.9	27.2	14.9
(8) lb of each fraction	290	136	74

Values for (1), (2), (3), and (4) pertain to batch 4.4, while (5), (6), (7), and (8) show the steps leading to the determination of the number of lb of each fraction comprising the 500 lbs of soil for batch 4.5.

The soil fractions on hand on the morning of 11 September 1958 were analyzed, and the following compositions were found.

<u>Bag Mark</u>	<u>Plus 100</u>	<u>Plus 200</u>	<u>Minus 200</u>
Red (R)	85 %	14 %	1 %
Yellow (Y)	10 %	69 %	21 %
Green (G)			100 %

It was then possible to put down the following equations:

$$\begin{aligned} \text{plus 100 mesh} &= 290 \text{ lb} = 0.85R + 0.101Y \\ \text{plus 200 mesh} &= 136 \text{ lb} = 0.14R + 0.69 Y \\ \text{minus 200 mesh} &= 74 \text{ lb} = 0.01R + 0.21 Y + G \end{aligned}$$

Solution as simultaneous equations gave the weights of soil that should be taken from each of the colored bags; thus,

Red (R)	324 lbs
Yellow(Y)	132 lbs
Green(G)	41 lbs

After contamination an analysis of the 500 lbs showed

	<u>Plus 100</u>	<u>Plus 200 mesh</u>	<u>Minus 200 mesh</u>
mass	62.9 %	22.2 %	1.43 %
activity	31.6 %	21.8 %	46.6 %

The values for mass were in good agreement with calculations ((7) above), and the values for activity were reasonable with those in (5) above. The inert soil addition to make 2000 lb was calculated to be (by method shown above)

Red (R)	187 lb
Yellow (Y)	500 lb
Green (G)	819 lb

After blending the inert soil into the mixer the final sample for batch 4.5 had the following composition

	<u>Plus 100 mesh</u>	<u>Plus 200 mesh</u>	<u>Minus 200 mesh</u>
mass	32.9 %	27.0 %	39.9 %
activity	23.2 %	18.4 %	58.2 %

This derivation of mass distribution from that calculated was undoubtedly caused by the addition of the inert soil, with the obvious inference that the inert soil did not conform to the analysis given above.

Figure 17 shows the distributions for batch 5.6 which was the last and best batch of synthetic fallout. These results were accomplished by the same methods described for batch 4.5, except with a much greater attention to every detail. Radiochemical assays were done in duplicate, the mixer was scrupulously cleaned, sieve analyses were made on thoroughly blended composite soil fractions, and the soil was precisely weighed.

The dispersal phase of the operation presented no major difficulties. As previously pointed out, the initial mass levels desired on the test surfaces were 10 grams/ft², 33 grams/ft² and 100 grams/ft². These levels were nominal mass levels which could represent three different fallout conditions that might be encountered during recovery operations following a land surface nuclear detonation. The three different levels were selected to allow the evaluation of the recovery procedures at various mass levels. No great amount of effort was placed in dispersing the synthetic fallout material at exactly these amounts. By the means described in Section 3.4.3, the radiation measurement obtained could be converted to mass, and thus it was only important to insure the uniform dispersal over each test area and not from test to test. In most cases this was accomplished. The largest difficulty encountered in the dispersal operation was the adverse effect of the wind. During the operation, tests had to be cancelled on three different occasions due to winds above 10 knots which would not allow uniform distribution of the synthetic fallout material on the test areas.

The conversion factor C, discussed in Section 3.4.3, was dependent upon the calibration factor K, for the mobile shielded gamma detector.

$$\text{Equations (6) - } M_0 = \frac{I_r}{C} \text{ g/ft}^2, \text{ and (7) - } M = \frac{R_r}{C} \text{ g/ft}^2$$

above are applicable only if K is a constant for all cases. A preliminary analysis indicated a considerable scatter in the data and, apparently, a variation of K with initial mass. Scatter in the data was to be expected and could be attributed to two major sources: wind effects and instrument errors. Moderate to heavy winds (5 - 15 knots) could change the contamination pattern between the time the pan samples were collected and the time the initial readings were taken, causing a discrepancy between g_1 (measured weight collected in pan) and I_r (egghead reading). Furthermore, on those days when the synthetic fallout contained disproportionately large amounts of radioactivity

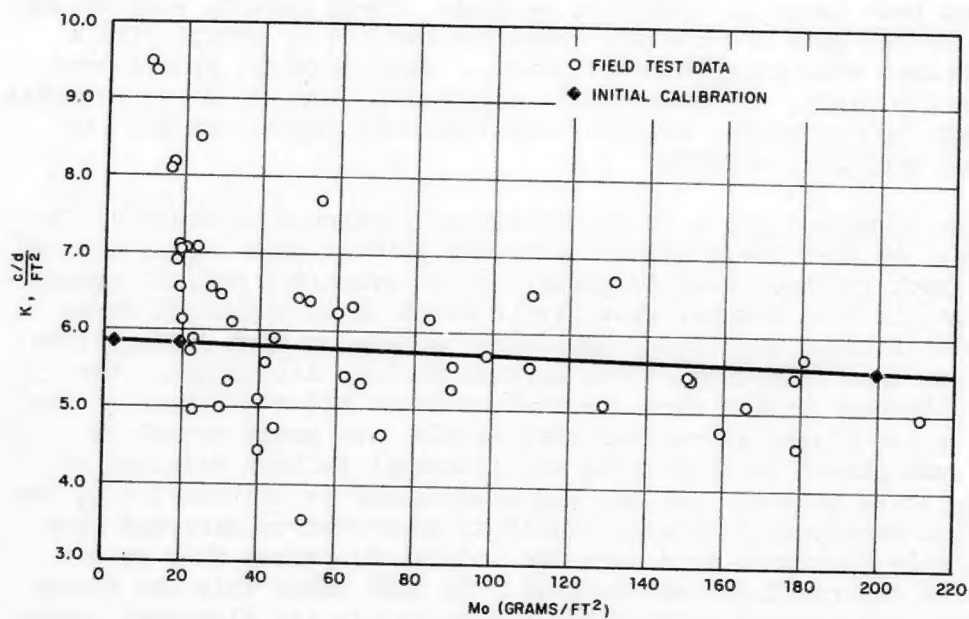


Fig. 16 Variation of K with Initial Mass Level on Asphaltic-Concrete Surfaces

on the small soil particles, any wind could cause fractionation during dispersal, resulting in anomalous results. The mobile shielded detector has an estimated inherent error in its output of $\pm 12.5\%$, which accounts for most of the instrument error. The 4-pi ion chamber with which specific activity was determined, has an inherent error of less than 2% including weighing errors. A standard estimate of error was run on five samples; the error in K ranged from $\pm 17.4\%$ to $\pm 34.3\%$. The primary source of error was in the weight of the pan sample; an important secondary source of error was the readings from the mobile shielded detector.

An inspection of the data for asphaltic concrete surfaces (Fig. 16) suggested that K might vary with M_0 . Although such a variation was not in keeping with theoretical considerations nor consistent with the results obtained from the initial calibration, also shown in Fig. 16, the trend in the data points was obvious enough to warrant further study. A linear regression on 21 points, selected on the basis of reliability of the data (low wind speed, uniform coverage with the contaminant), was made, which indicated a slope of 1.2×10^{-6} c/d/g; the slope of line K, the initial calibration, was 1.5×10^{-7} c/d/g, which slope could be attributed to the self shielding in the contaminant itself. Several statistical tests were applied to the results of the linear regression. Not unexpectedly a large degree of uncertainty was indicated, with the limits of the slope being $+4.7 \times 10^{-6}$ to -1.5×10^{-6} . It was concluded that no significant variation of K with M_0 occurs and that, for practical purposes, a simple average would give the best value for K.

Selection of K_0 for Various Surfaces

A K of constant value, denoted K_0 , was established, for each of the five different surfaces tested by a simple average of all suitable values. Individual K values were eliminated from the averages if:

- (a) The ratio $(c_1/m)/g$ varied more than $\pm 5\%$ for more than 20 % of the samples in each test.
- (b) The ratio c/d varied more than $\pm 5\%$ for more than 20 % of the samples.
- (c) Specific activity for an individual test varied by more than 10 % from other values for specific activity for the same batch of contaminant.
- (d) The wind speed during the test exceeded 9 knots.

In addition the standard deviations for I_r for each test was determined; individual values of I_r which exceeded three standard deviations were eliminated from the averages.

The K_o values established are as follows:

Surface	No. of Tests	% of Total	K_o c/d(ft ²)	K
Asphaltic Concrete	41	82.0	5.870	0.838
Portland Cement	12	92.3	6.290	1.349
Green Grass	3	100.0	5.730	-
Tilled Soil	4	80.0	4.789	-
Dry Soil	5	100.0	5.750	-

The appropriate values of K were inserted in equation (5) to obtain a new value for the conversion factor, C. This derived conversion factor C, when applied in equation (6), $M_o = I_r/c$ and (7) $M = R_r/c$ enabled the results obtained from the decontamination tests to be expressed in mass units of g/ft². This technique was the basis for determining decontamination effectiveness and the results are presented in Volumes II, III and IV of this series of reports.

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For the Scientific Director

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